High Resolution Iterative Image Reconstruction from X-ray Speckle Patterns

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Abstract

- We have reconstructed images of two-dimensional non-periodic phase objects from their experimental coherent soft-X ray transmission diffraction patterns using the iterative Hybrid Input-Output (HiO) algorithm [1,2]. Agreement between the reconstruction and SEM images of the same object was obtained by applying a sign constraint and a stabilizing procedure. Low resolution images were used to provide a disjoint support. Resolution in these images of 50nm diameter gold balls is estimated to be about 10nm.

[1] J. R. Fienup, J. Opt. Soc. Am. A 4, 118 (1987) and references therein.

[2] J. Miao, P. Charalambous, J. Kirz, and D. Sayre, Nature (London) 400, 342 (1999).

Why Iterative HiO?

- No lenses are needed, no aberrations, the only limit to resolution is wavelength
- No crystal needed, non-periodic sample OK
- 3D tomography possible.
- Opens up possibility of imaging with radiations for which no lenses exist

Phase Problem

Problem: Missing phase information in k-space, half of the information.

Solution: ‘Over’ sampling. In real space, the object is known to be 0 outside a support s.

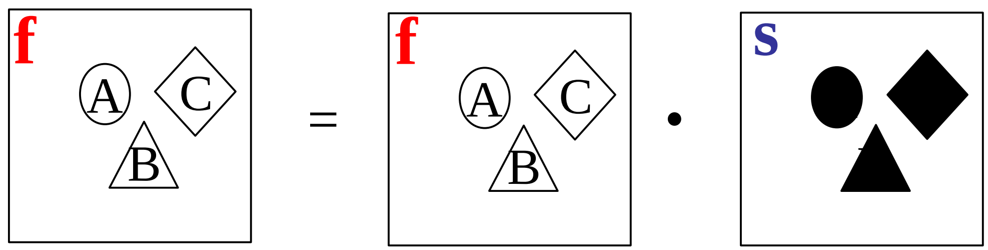
If S=FFT(s) is non-null in at least 2X2 pixels, the self consistent equations bound each pixels with enough constraints

Self consistent equations

Unknown inside the support s

Known

Real space $\boldsymbol{f=f\cdot s}$



approximation

FFT ↕ IFFT

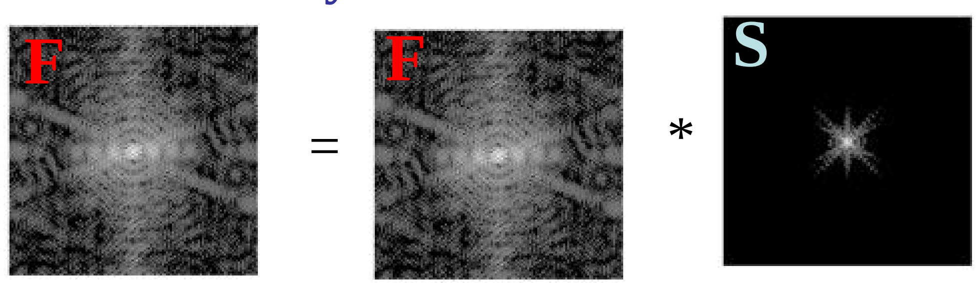
Unknown phase

known intensity

FFT ↕ IFFT

Known

K-space $\boldsymbol{F=F*S}$

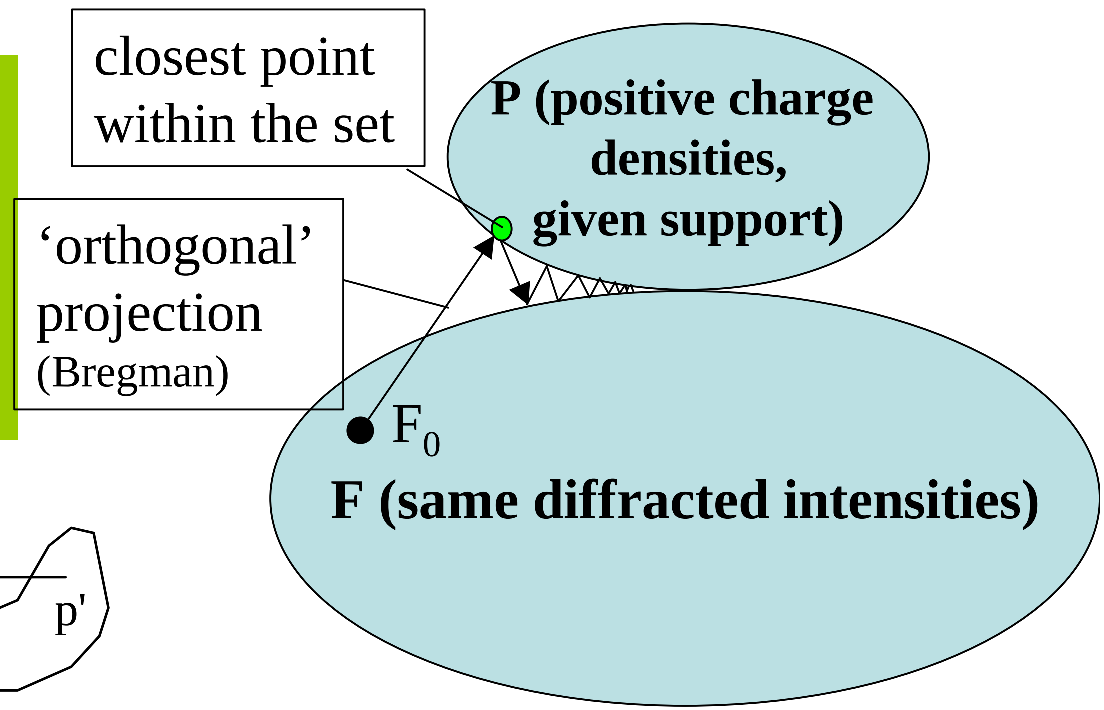


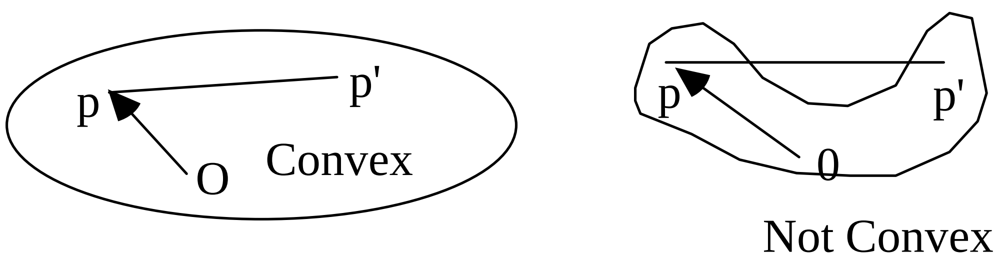
Phase Solution

<u>Uniqueness.</u>
Eqns no longer impossible to solve. But non-linear !
The non-linear eqns may not be independant. Barakat, Newsam (1984) show multiple solns for 2-D complex are "pathologically rare". (Analyticity, with support.)

<u>Global Optimization.</u>
2. Find metric c^2 which measures similarity of members in each set.
3. Iterate between most similar elements of each set to find solution.

<u>Examples of convex constraints</u>: Symmetry, Positivity, Atomicity, Compact support, Non-convex: Fourier modulus, phase object (phase must move on circle with thickness).





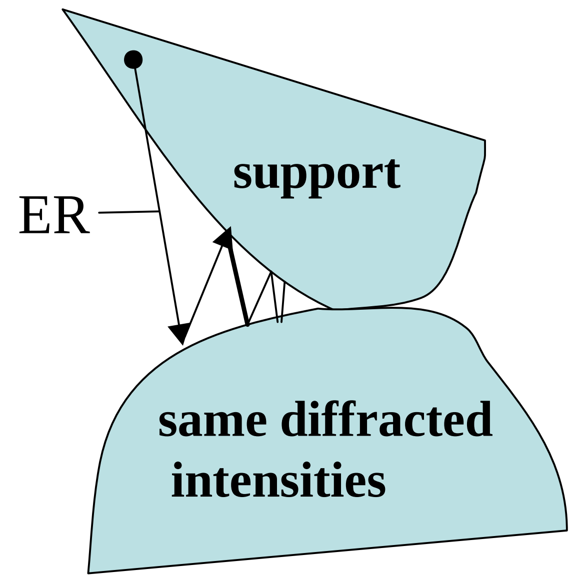
Error Reduction (ER)

Self consistent equations

Real space
$f = f \cdot s$

FFT ↑↓ IFFT

K-space
$F = F * S$



Known:

- **k-space amplitude:**
  $\lvert F \rvert$
- **Support *s* in real space:**
  f=0 for s=0

Unknown:

- **Phase in k-space**
  j
- **Image inside the support**
  ***f*=? For *s*=1**

	j =rand	known info	
K-space	$F = \lvert F \rvert e^{ij}$	$j = j'$	$\lvert F' \rvert e^{ij'}$
	FFT		IFFT
Real space	f	$f' = f\,s$	f'

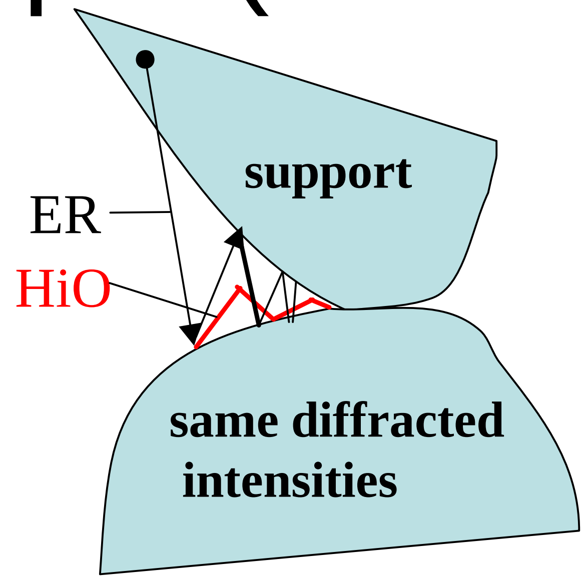
Hybrid Input Output (Fienup)

Error reduction:

$$f = f \cdot s$$

HIO

$$f' = f \cdot s + (f_{old} - b\, f) \cdot (1 - s)$$

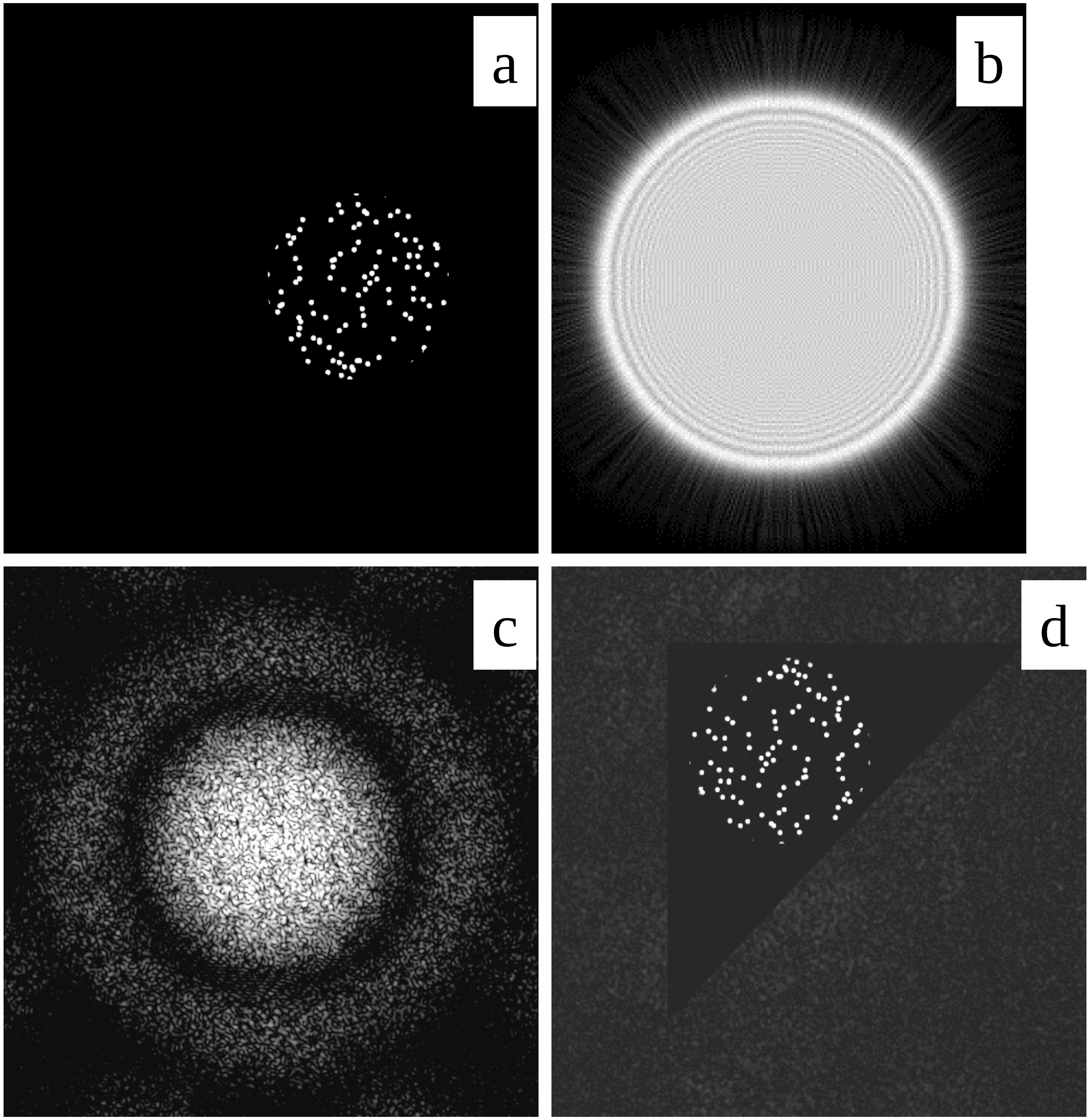


The step outside the support is relaxed, with a parameter 0.5<β<1. β=1 corresponds to a full step.

The idea is derived from the conjugate gradient method.

	j =rand	known info	
K-space	$F=\lvert F\rvert e^{ij}$	$j = j'$	$\lvert F'\rvert e^{ij}$
	FFT		IFFT
Real space	f	*HIO*	f'

Simulation example (real object)



A. Object - pinholes, 10nm diam.
B. Incident 2.5nm soft Xray wavefield from 1 micron aperture at 200nm.
C, Speckle pattern.
D. Reconstruction,150 itns (inverted)

Triangular support used (just visible) in d.

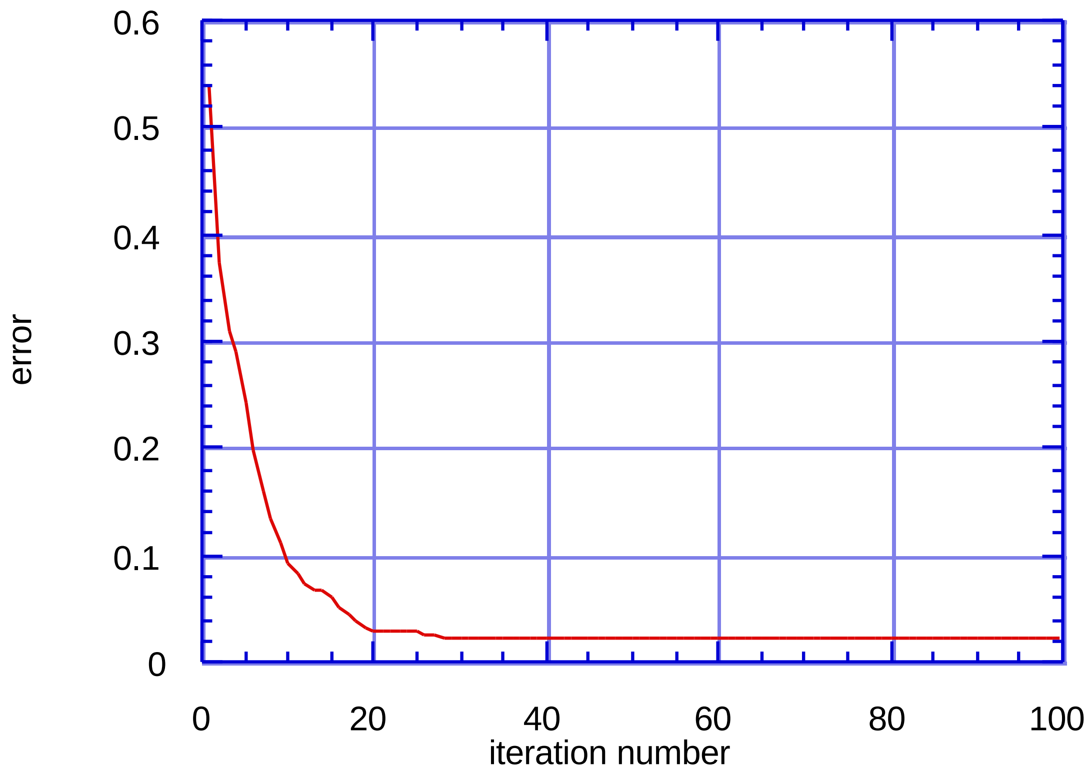
Phase problem solved for non-periodic object !

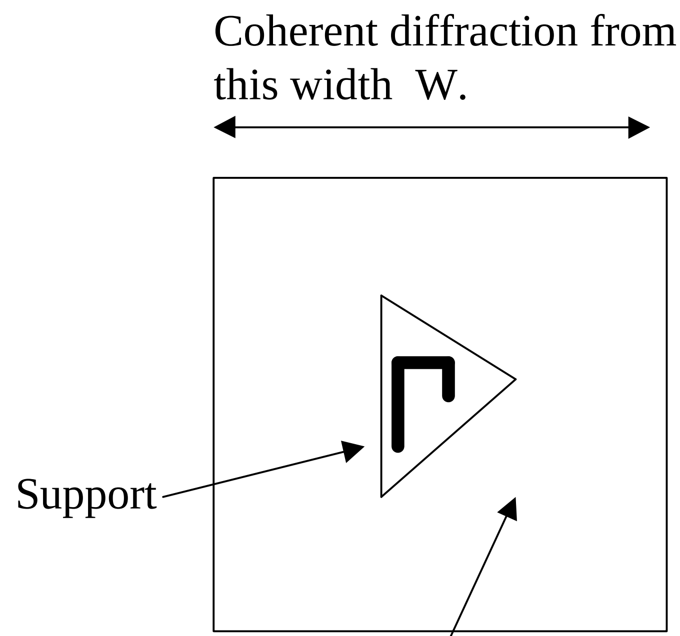
Note: convergence is independent of the particular set of randomly chosen phases we start with.

U. Weierstall et al
Ultramic. 90, p.171 (2002).

Decline of error with iteration for typical simulated real object.

U. Weierstall et al
Ultramic. 90, p.171 (2002).

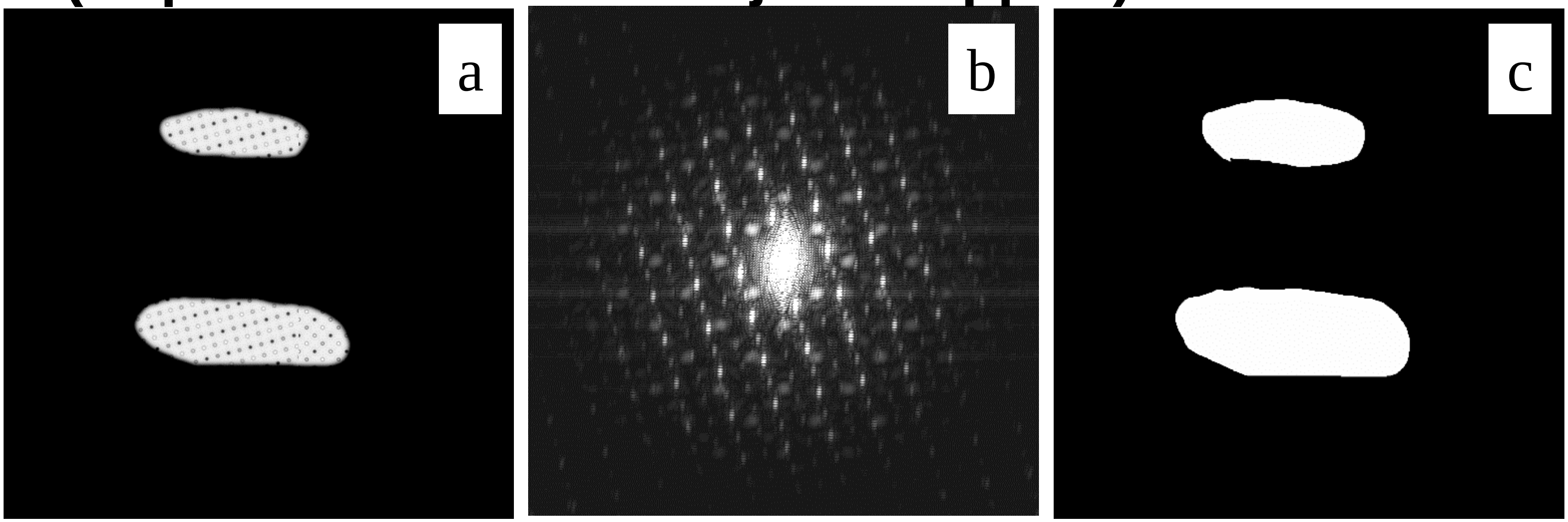


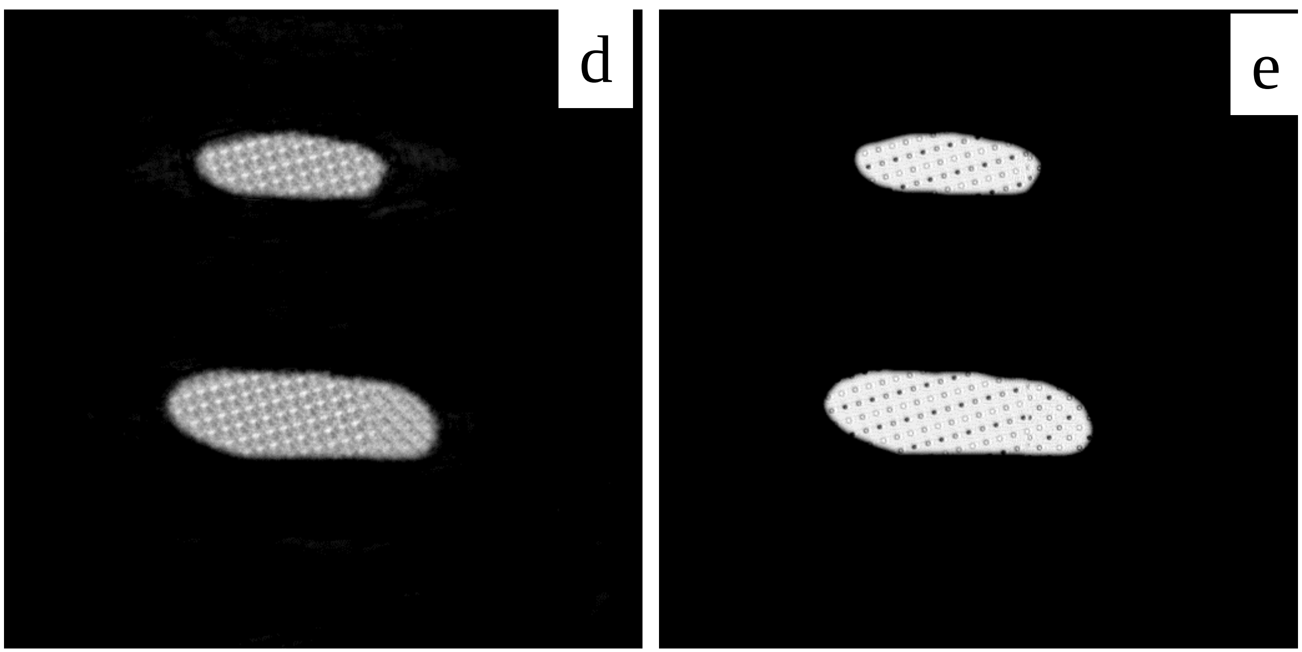


Error is goodness of fit index for known zero region outside triangle.
Rely on uniqueness theorem to establish that if i) image is correct (zero) in known region, and ii) Fourier modulus is correct (measured), then image must be correct within unknown support region.

HiO simulation for complex object (requires “two-hole” disjoint support).

U. Weierstall et al
Ultramic. 90, p.171 (2002).





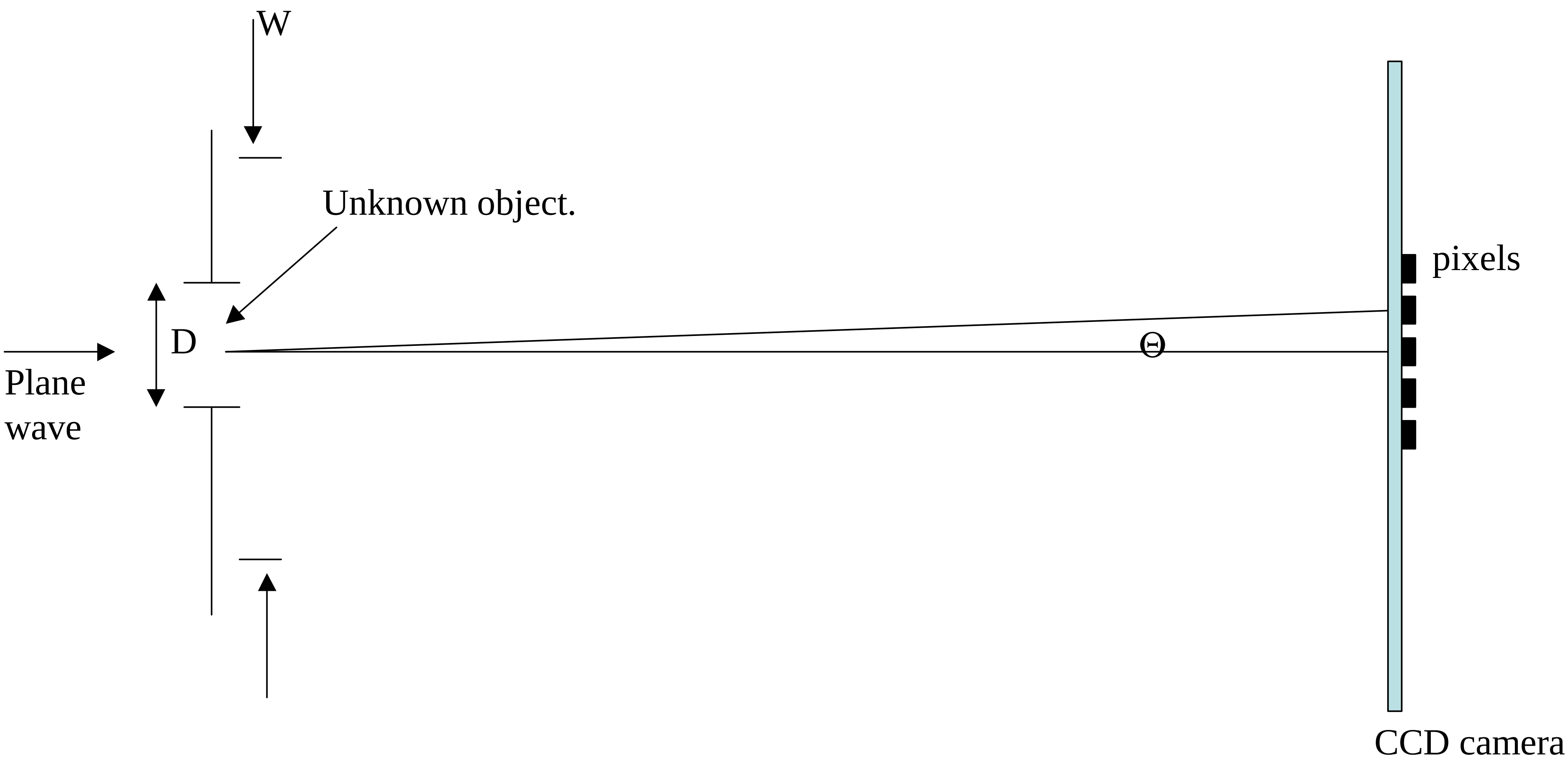
a. Simulated object: electron wavefield at thin silicon crystal
b. Diffraction pattern. Wavelength 0.025 Angstroms.
c. Support assumed,bigger than holes
d. reconstruction after 100 itns.
e. Reconstruction after 3000 itns.

Note: Because support does not exactly match holes, convergence is slow.

cf FT Holography. No beam-stop problem

“Complex object” means phase shift exceeds $\pi/2$.

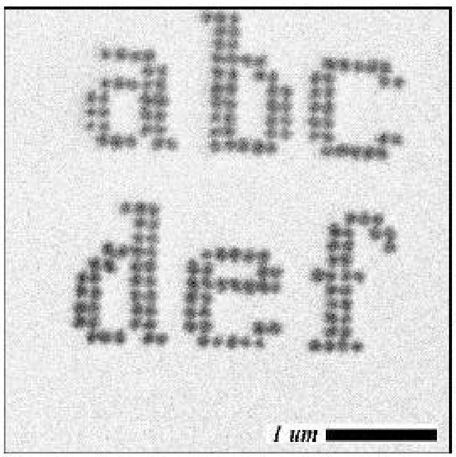
Experimental requirements for lensless imaging.



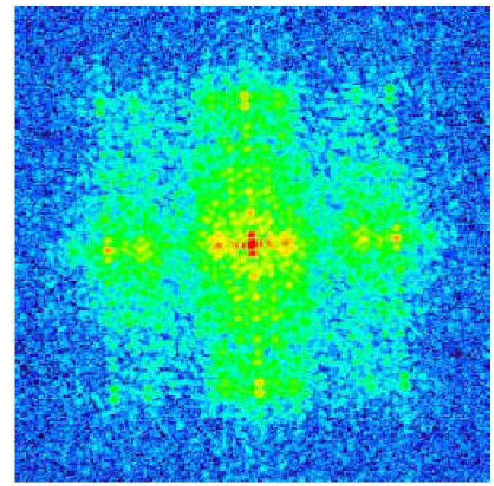
1. **Lateral coherence** width must exceed W > 2D.
2. **Temporal coherence** across W to resolution required. $E/\Delta E$ = linear number of pixels.
3. Diffract from width W larger than known size D of object. Hence $W = \lambda / \Theta > 2D$ fixes **working distance**. (First order "Bragg beam" at Θ corresponds to a periodicity larger than object, ensuring oversampling The number of pixels fixes largest scattering angle and hence resolution).

Soft x-ray imaging (Miao et al. Nature 1999)

SEM Image

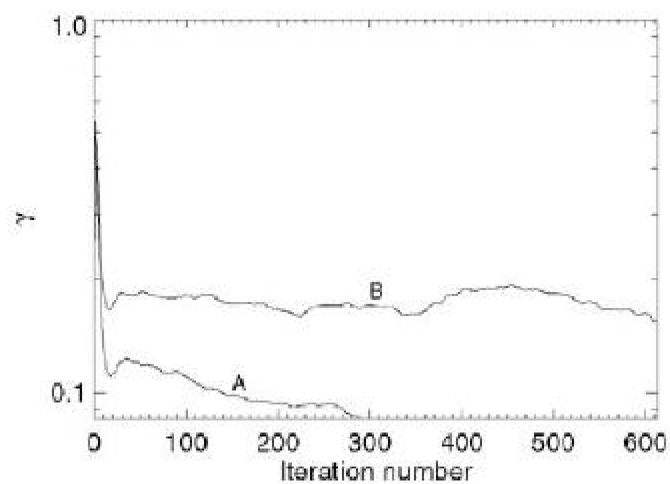


The Diffraction Pattern



A Reconstructed Image (after 400 iterations)

The convergence of the reconstructions



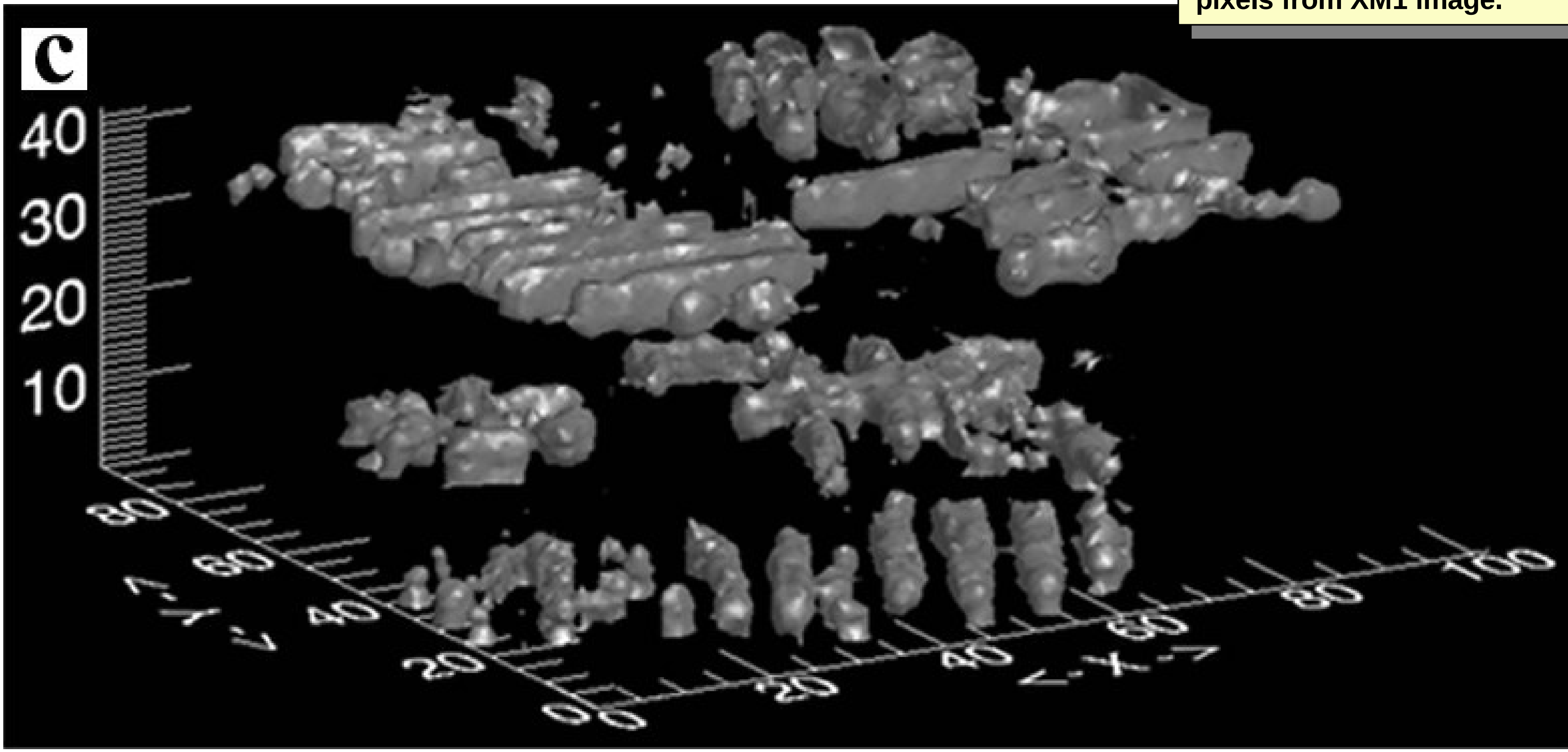
3D soft x-rays: Tomographic HiO

J. Miao, T. Ishikawa, B. Johnson, E. H. Anderson, B. Lai, and K. O. Hodgson, Phys. Rev. Lett. 89,

1. Miaow's 8nm res. is from diffraction pattern.

2. XM1 "low res" image is almost as good.

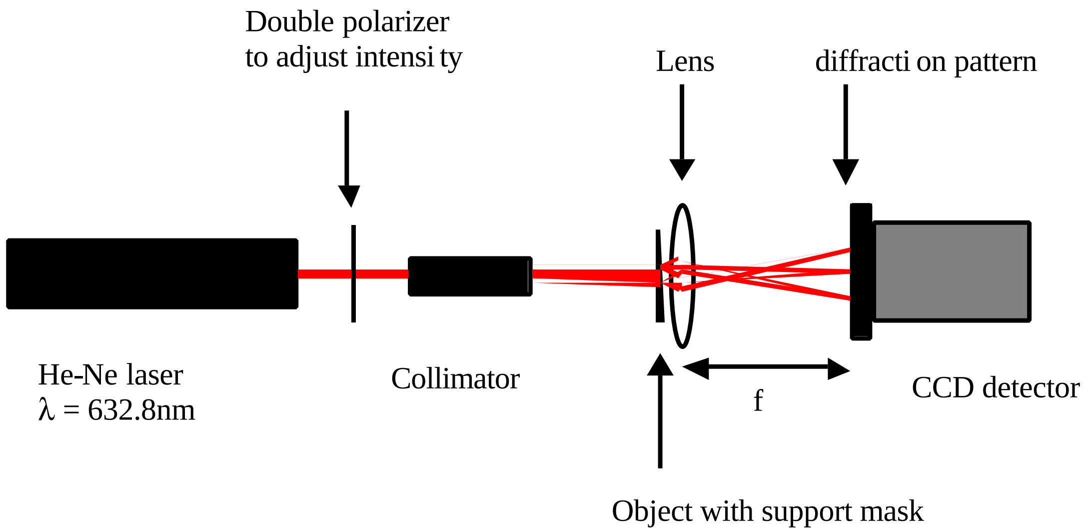
3. Miaow uses only inner 60 pixels from XM1 image.

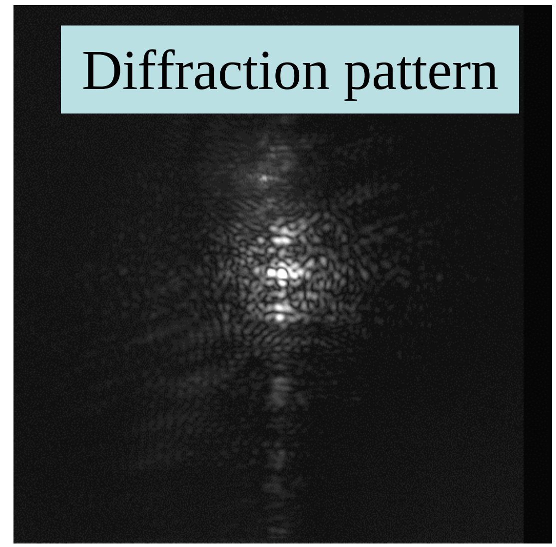


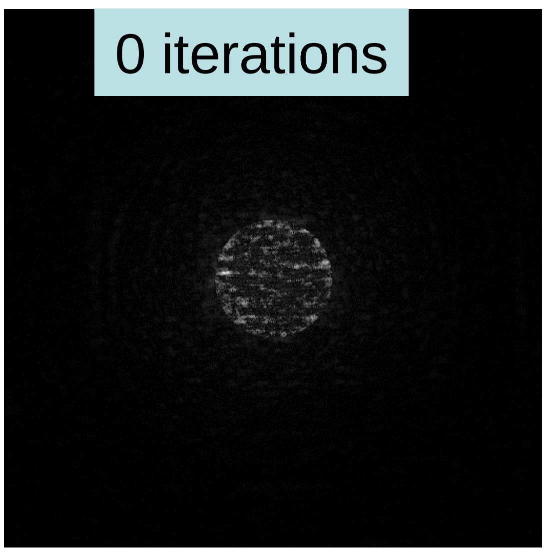
HiO reconstruction in 3-D from coherent 2 Angstrom XRD patterns. Resolution is 55nm.
Spring-8. Double xtal Si mono., undulator. Lithographed Ni multilayer structure. **Low res image from ALS** XM1. (innermost 60 X 60 pixels). Recording time 20 mins for each of 31 patterns needed. Computing power limits data collection to 55nm resolution in 3D, could be 28nm. (2D res. is 8nm).

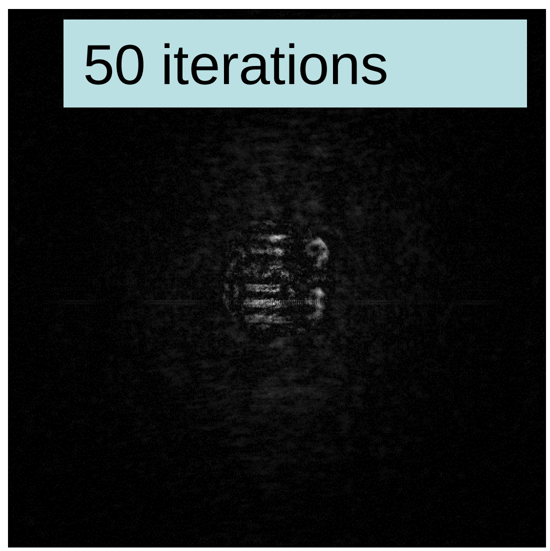
Visible light (2D)

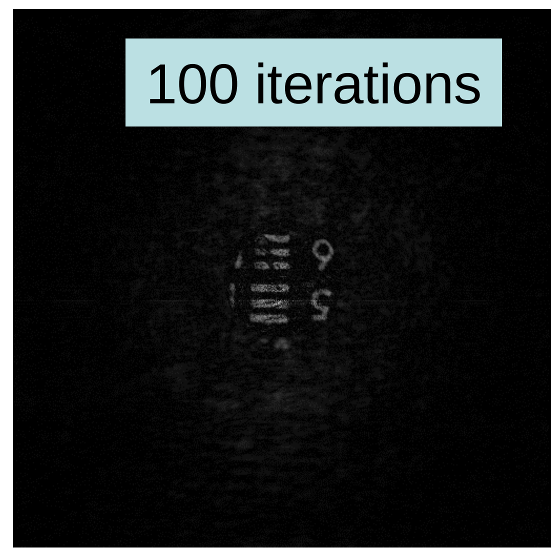
J. Spence et al. Phil Trans. "Thomas Young" issue. 2001.
U. Weierstall et al. Ultramic. 90, p.171 (2002).







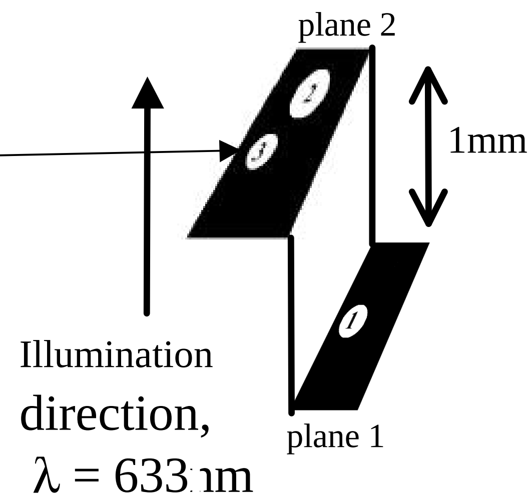


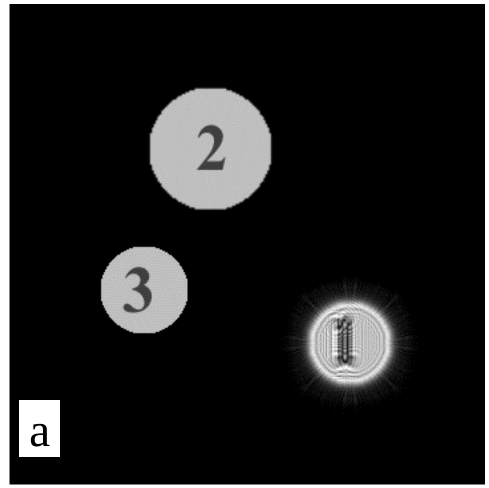


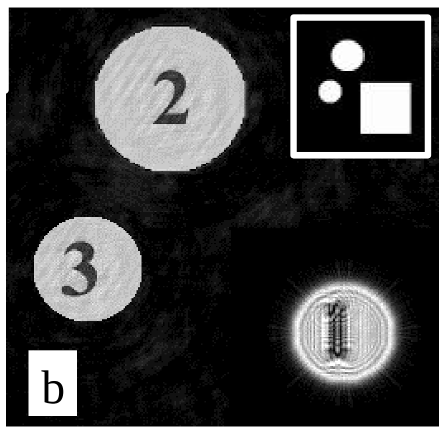
Visible light (3D)

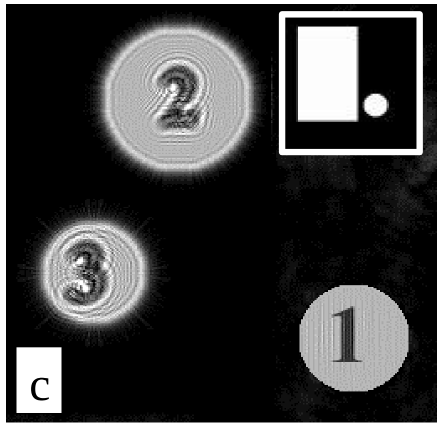
U. Weierstall et al. Ultramic. 90, p.171 (2002).
J. Spence et al. Phil Trans. "Thomas Young" issue. 2001.

300µm holes
with complex number
objects:
transparency 20%
phaseshift 2 rads in
letter



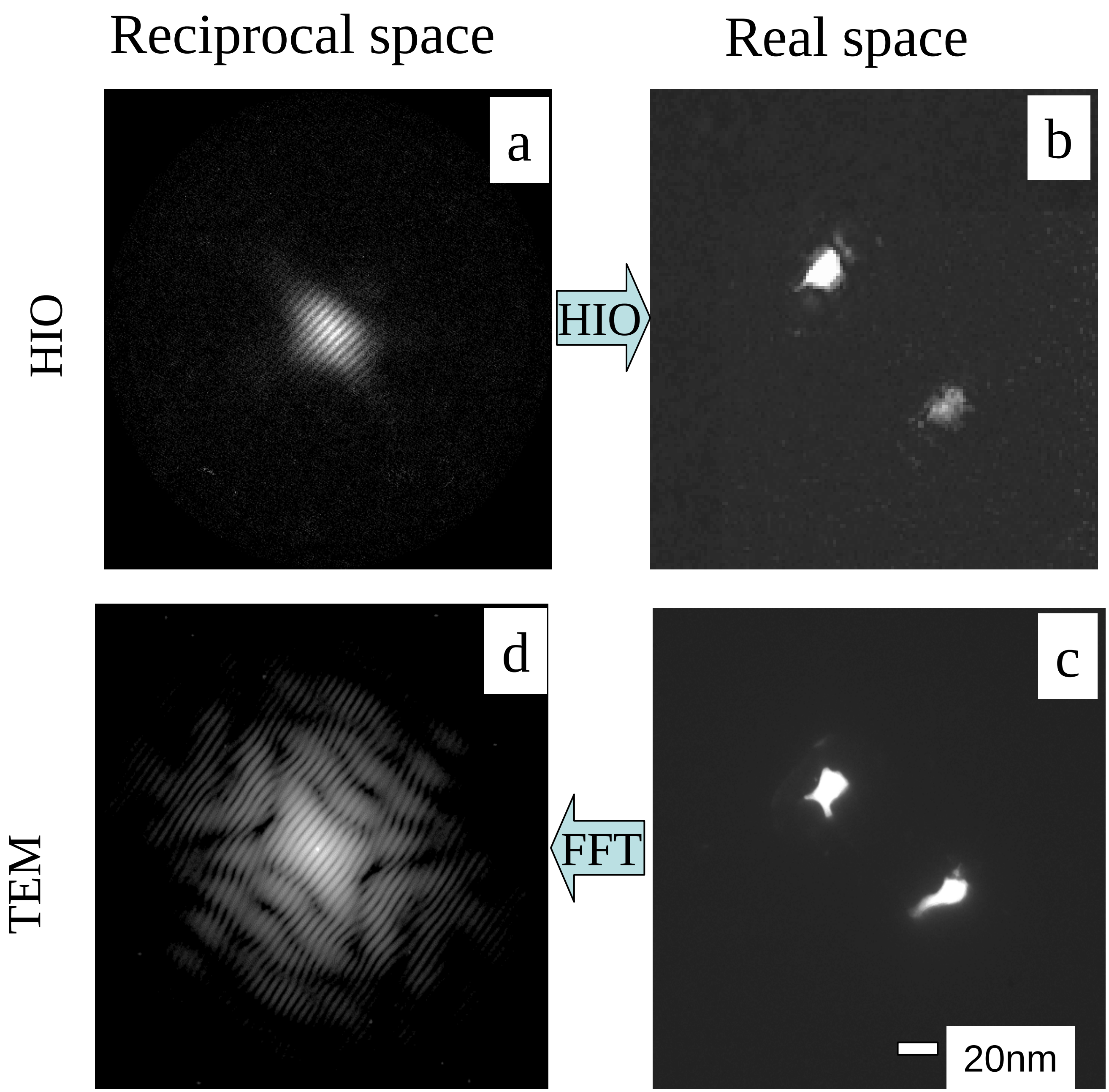






Coherent electrons

J. Spence et al. Phil Trans. "Thomas Young" issue. 2001.



a) Diffraction pattern of two-hole object in c).
b) Image reconstructed by HiO after 100 iterations.
c) TEM image of object.
d) FT of TEM image.

Conditions:
Field-emission gun, 40 kV,
Lithographed object
Philips CM200 CCD camera.

Note that opaque support solves beam-stop problem !

Weierstall et al Ultramic 90, p. 171 (2002).

Summary: HiO for 2-D, Opaque Supports

	General complex object Ψ=Aexp(iϕ)	Pure strong phase object Ψ=exp(iϕ(r))	Pure "weak" phase object: $\phi<\pi/2$	Real (amplitude) object	weak-phase object with absorption
Known two or more hole physical support	ca, *(Laser exp.: pin, cheek cells)*	ca, p *(Laser exp.: mica)*	rip, ca	m, rip, ca	rip, ca
Known one hole phys. support	ca, only for certain support shapes (e.g. triangle with sharp edges)	ca, slow convergence, p works better	rip, ca	m, rip, ca *(Laser exp.: numbers)*	rip, ca
Unknown two or more hole phys. support	-	-	rip	m *(TEM exp.: holes)* *(Laser exp.: numbers)* *(planned: X-ray, mica pinholes)*	rip *(Miao X-ray exp., Transparent support)*
Unknown one hole phys. support	-	-	(rip)	(m, rip)	(rip)

ca - complex algorithm

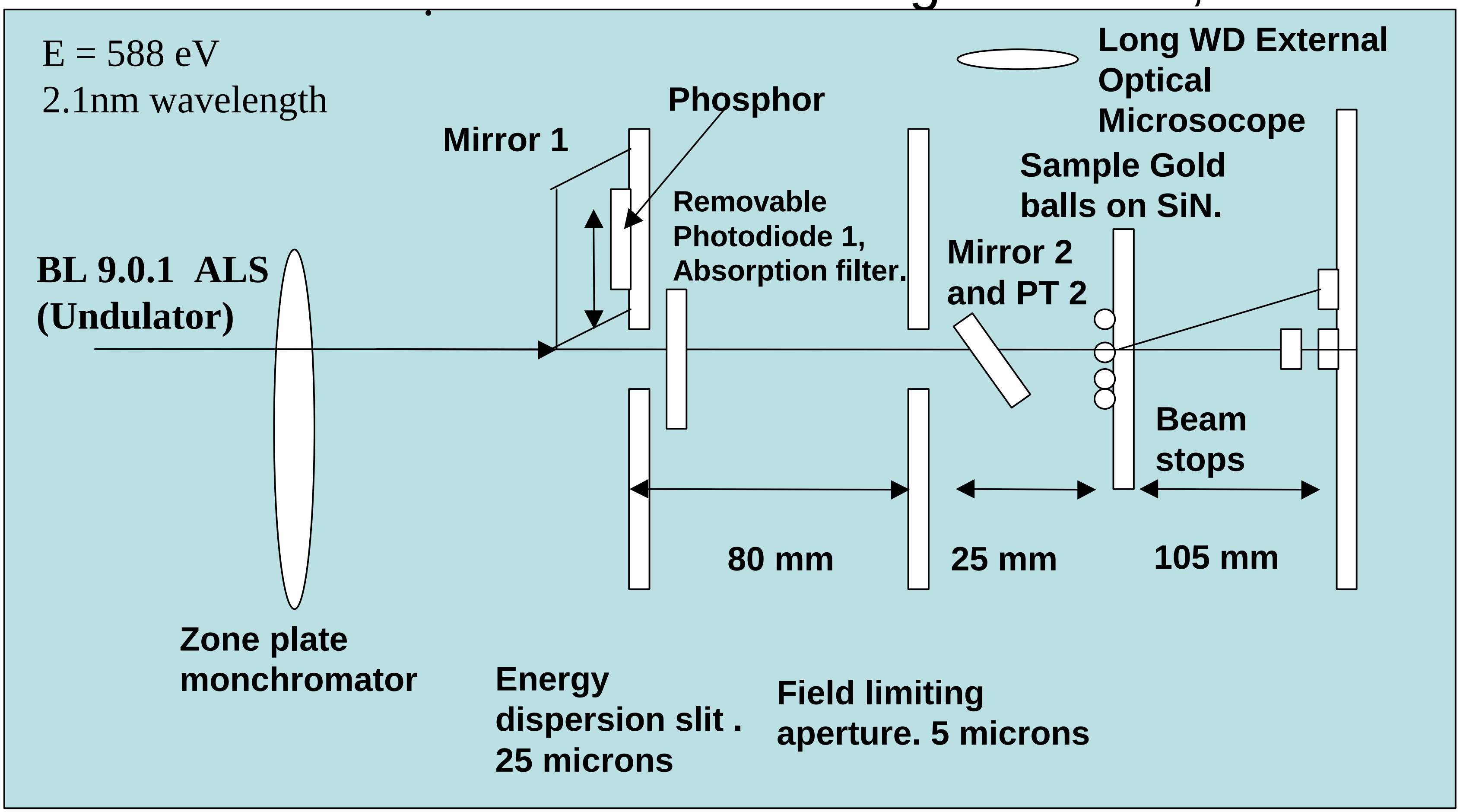
rip - positivity of ReF, ImF in real space

m - modulus constraint on F in real space

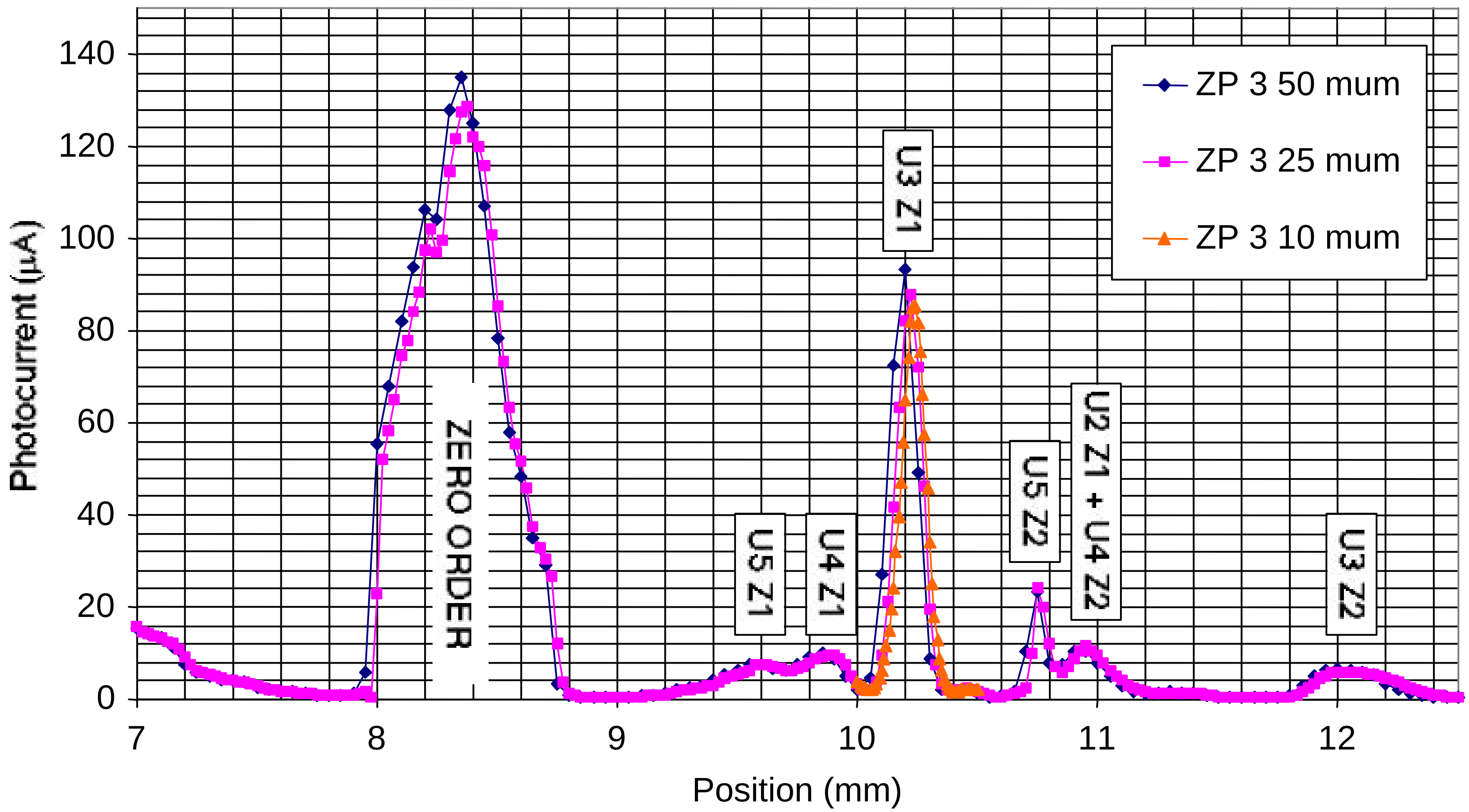
p - phase constraint on F, set modulus = 1 in real space.

Experiment (soft X-rays)

Layout of the diffraction chamber used for this experiment at BL 9.0.1 at Advanced Light Source, LBL



ZONE PLATE POSITIVE-ORDER SPECTRUM

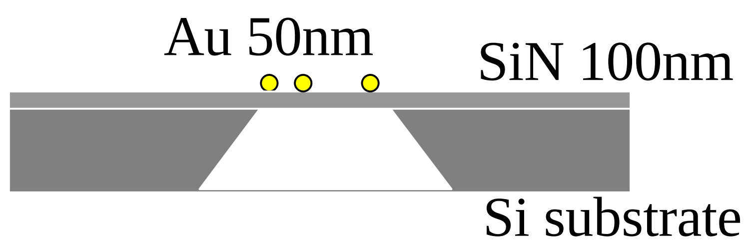


Peak width contributions	• Diffraction: 2.3 µm • Geometrical image: 2.3 µm • Source bandwidth:20 µm • Slit size: 25 µm • Defocus: 90 µm	• Overall calculated value: 96 µm • Measured value: 110 µm

Sample

- Sample: 50 nm gold balls randomly distributed on SiN window (~100nm thickness and $2\times2\ \mu m^2$)

Side view of sample

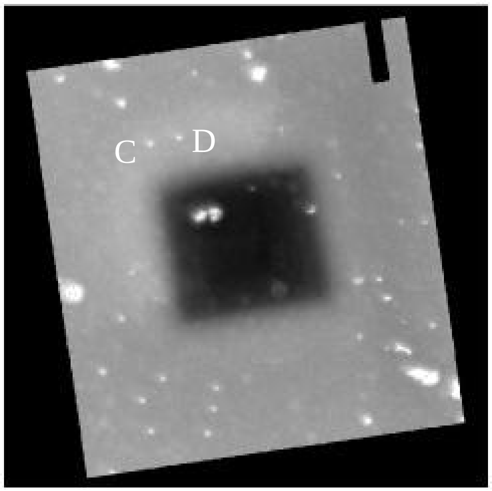


- Wavelength 2.1nm (588 eV)
- Detector: 1024×1024 Princeton back-illuminated CCD

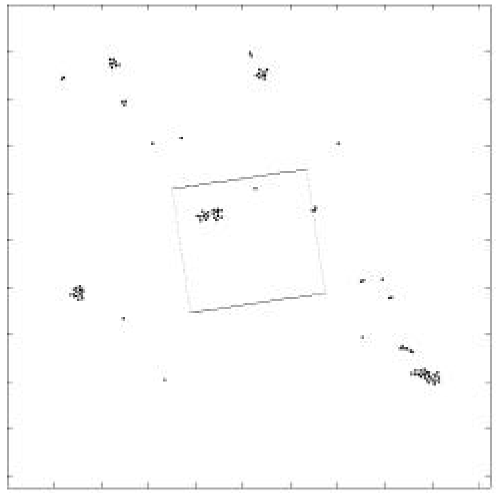
SMALL WINDOWS

- Small SiN windows of around 2×2 μm have solved many of our problems - eg how to make an isolated sample with known support.
- Do not produce excessive stray X-rays due to edge scattering like laser-drilled metal pinholes - the edge is too short to have much scattering power
- Their diffraction pattern is visible outside the beam-stop from which the dimensions of the window can be found.
- They provide sample isolation inside a known and tight support compatible with oversampling. How to make an isolated sample of submicron dimensions ?
- They reduce strength of direct beam sufficiently that beam stop can be removed, allowing central region of pattern to be recorded with filters (No missing data)
- Stray light is reduced in proportion to window size
- Coherence width of beam need be only equal to window width.
- They simplify the problem of finding the isolated sample, otherwise almost impossible. (2D search over 0.1mm square area for submicron object).

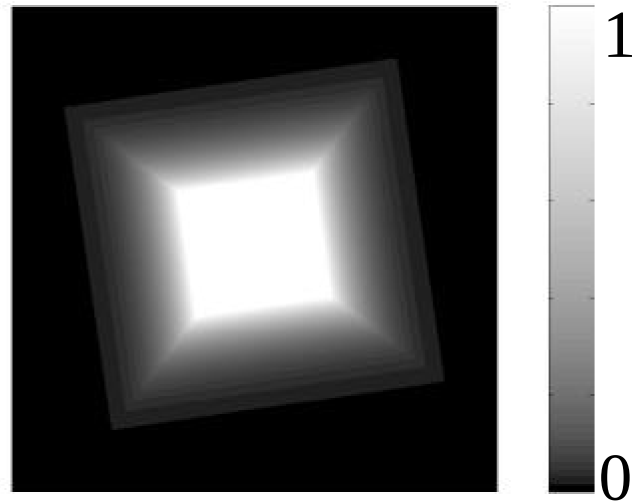
Sample



(a) SEM image of sample which produced the diffraction pattern in fig. 2. The dark square in the center is the 100nm-thick silicon nitride membrane (see also fig. 5(c)). See fig. 4(a) for correspondence of C and D.

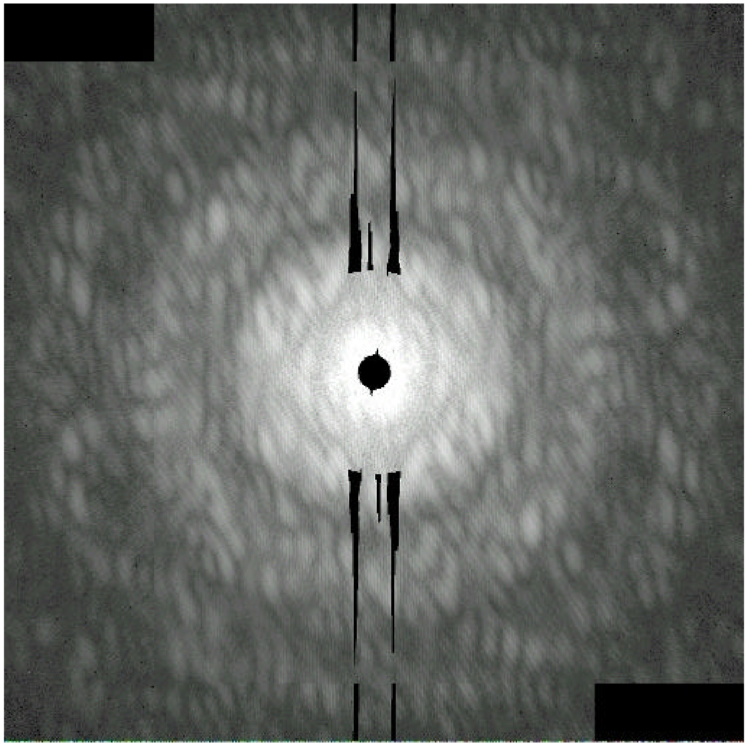


(b) Positions of balls extracted from SEM image in fig. 5(a). The position of the window is shown approximately.

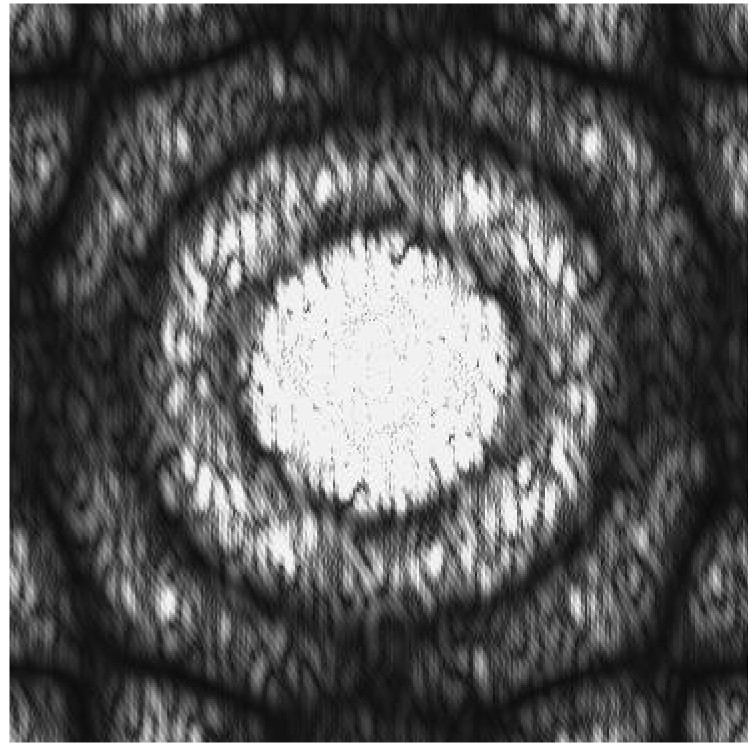


Simulation of SiN window and surrounding Si wedge transmission which produce the central diffraction pattern shown in fig. 3(b).

Diffraction pattern

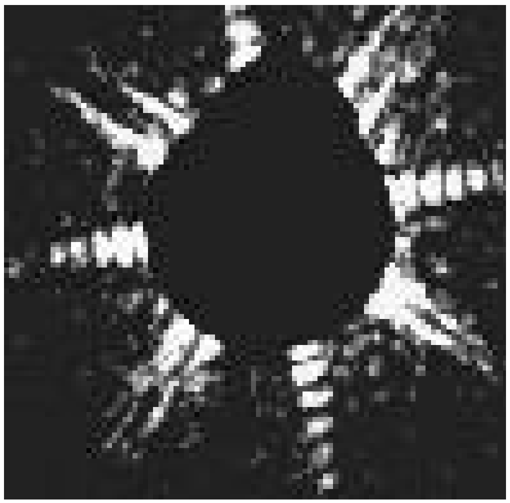


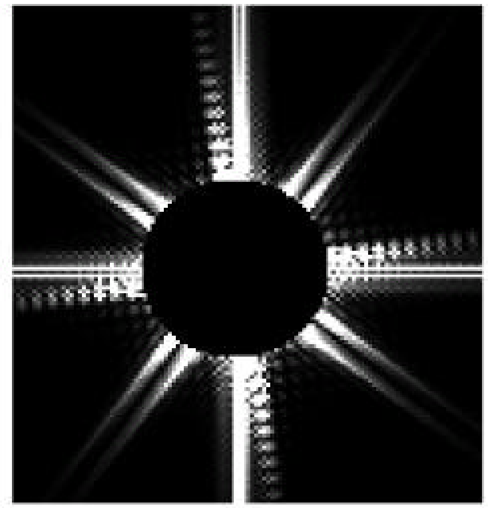
- **Fig. 2(a) Experimental diffraction pattern from sample Au5010. The pattern has been averaged by inversion, and contains artifacts from camera readout. The dark region in the center and edge and in between are missing data.**



- **Fig. 2(b) Simulated pattern based on ball positions obtained from an SEM image of the same object. SEM image is shown in figure 5(a)**

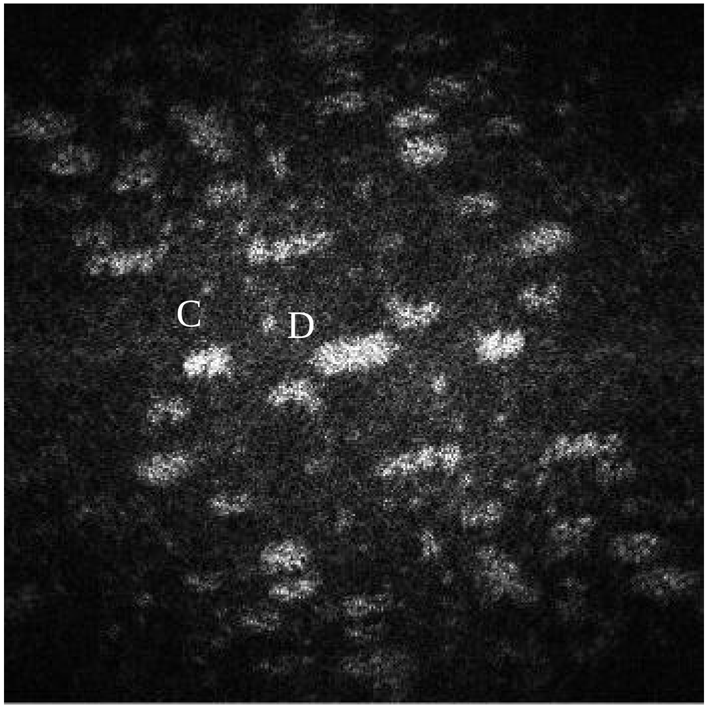
Diffraction pattern

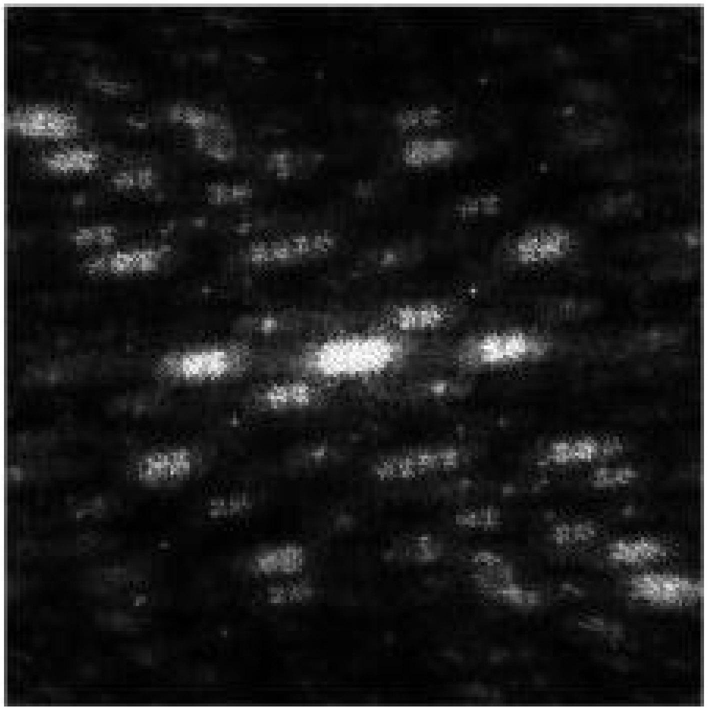




- **Fig. 3(a) Central region of a pattern similar to fig. 1, showing 1mm beam stop bead and the sinc-function like pattern from the silicon nitride window. Additional streaks are seen which arise from the valleys running from the corners of the SiN windows.**
- **Fig. 3(b) Simulated soft X-ray diffraction pattern from silicon nitride window (see fig. 5(c)) with 54 degree wedge-shaped borders. The pattern is in good agreement with Fig. 3(a).**

Autocorrelation





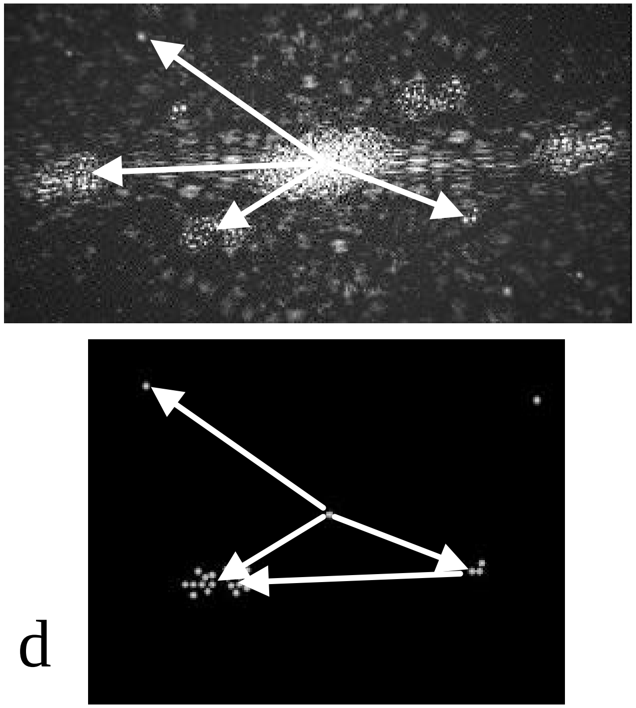
- Fig. 4 (a) Fourier transform of the intensity distribution shown in Fig.1. This is the autocorrelation (Patterson) function of the object. This is a map of all interball vectors with a common origin. Note single ball features at C and D, which correspond to the balls in the SEM image fig. 5 marked at C and D.
- Fig. 4(b) As a comparison, autocorrelation function obtained from SEM image (fig. 5(b)) is shown.

Autocorrelation

Enlarged portion of autocorr fn, showing real-space images of several clusters as formed by convolution with one isolated ball. Fig. 4(d) The real-space structure obtained from the SEM image, indicating the inter-ball vectors identified in fig. 4(c).

c

d



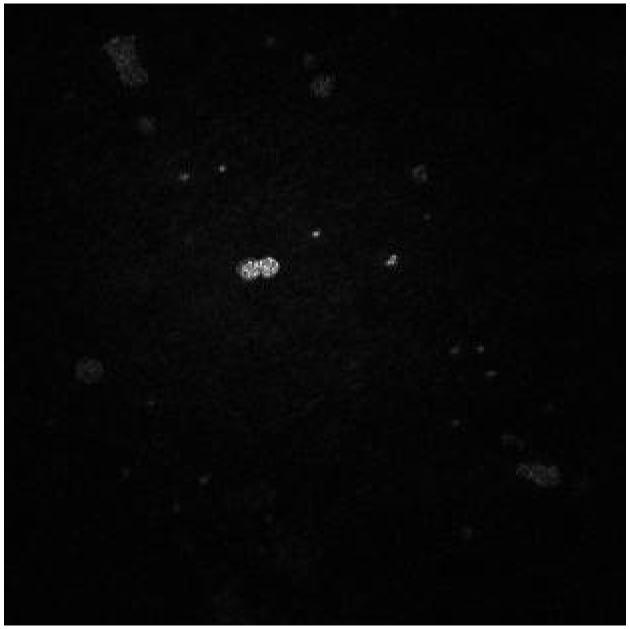
HiO+ for Reconstruction

- A slight change of the basic HiO has improved the reconstruction process when missing data is present (eg due to beam stop):

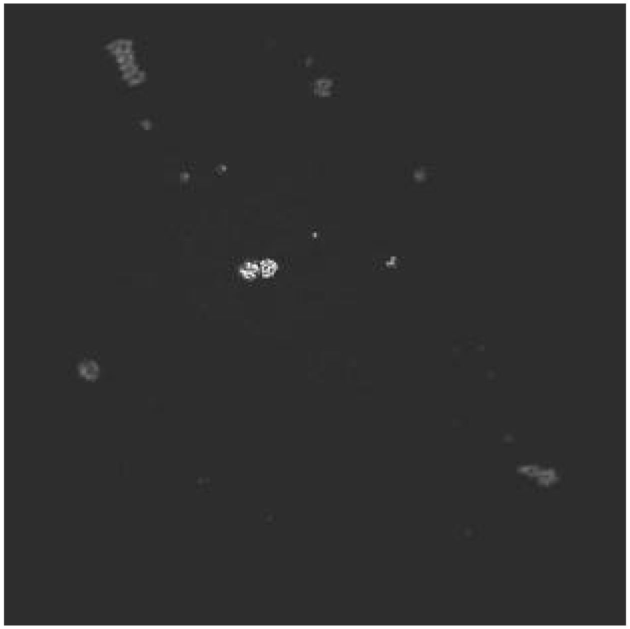
In step 2, instead of replacing |F’(u)| with full |F|, we replace |F'| only with known data, while allowing the missing data region to float. The idea is to treat the missing data as unknown information just like the missing phase information.
This is a general modification to HiO, doesn’t depends on the specific problem.

- Other constraints also greatly help the reconstruction process such as:
- **a). Positivity constraint: in our case, the gold ball is a phase object, the maximum phase shift q is less than p/2. Thus the real and imaginary part of the transmissivity exp(i q) of ball are both positive**
- **b). A unit modulus constrain may be applied to a pure phase object.**
- **c) A binary object constraint allows only two complex object values.**

Reconstruction results

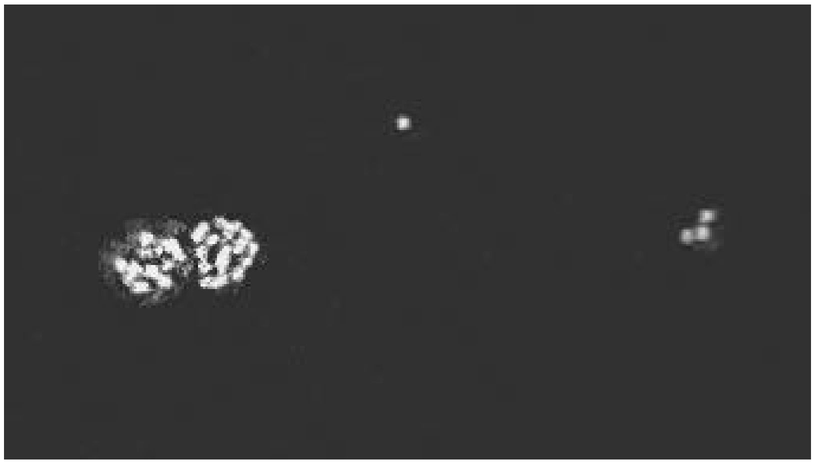


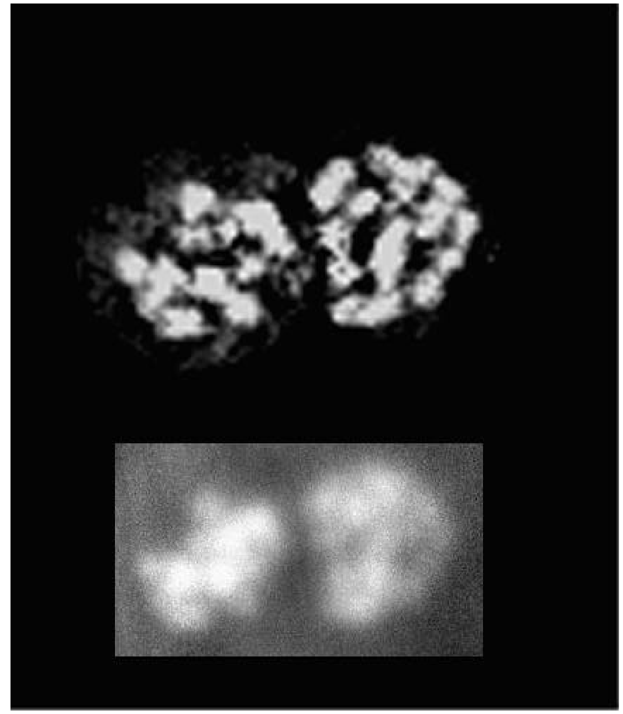
- **Fig. 6(a) Result of 420 iteration of HiO algorithm applied to experimental data of fig. 2(a) wihout using sign constraint. Support consists of boundaries drawn around clusters in SEM image. Note how balls outside window on Si wedges appear darker.**



- **Fig. 6(b) A better reconstruction of the same experimental data when unknown data are left floating, a positive sign constraint is used on gold balls, with 150 iterations. The same support is used. The HiO extracts internal detail (see fig 8).**

Reconstruction results





- **Fig. 7 Enlargement of inner clusters shown in fig. 6(b) , showing internal detail.**

- **Fig. 8 Enlargement of inner cluster compared with SEM image (lower).**
- **The ball diameter is 50nm, the Xray wavelength is 2.1nm.**

Reconstruction with other constraints

- We failed to reconstruct the image using only the positive sign constraint and without the stabilizing procedure. We note that the positive sign constraint works well alone with simulated data. Its failure with experimental data shows the insufficiency of this constraint with noisy data.
- Other constraints (eg modulus, binary object constraint) were also tried without success.
- Further attempts to model the Si wedge (which is partially transparent to our soft X-rays) and apply it as a constraint to the reconstruction are in progress. This will be useful if no balls lie on the partially transparent regions of silicon.
- Gold index of refraction constrain is being tried

Where is this technique going ?

1. **Other methods**:
   Cryomicroscopy (Baumeister): whole cells at 4nm res. in 700nm thick ice. Main problem: damage from many tilts. Cell size 0.5 micron.

   XM1 1-5 (?) μm thick unstained protein in 5 μm thick ice. Res. 25nm

   HVEM ultramicrotome tomography. Stained. 5 μm. 2nm res. Tedious!

   Use HiO to phase FEL data. Pulsed XRD from individual molecules. Tomographic SEM. Paul Midgely's movie.

2. **The challenge**: Find the atomic coordinates of every atom in a single inorganic nanostructure. Use DM and HiO with harder X-rays. Need...

Summary.

•**simulations** work perfectly with noise to solve the phase problem for non-periodic objects. For real object (X-ray diffraction) requires only a rough estimate of the size of the object. HiO works **better** in **3-D** (tomography). Phase shift across one voxel is always small.

•HiO applied to **laser-light** experimental data works well.

•HiO applied to coherent **electron** diffraction gives 1nm resolution. But use of imaging mode on TEM gives 0.1 nm resolution ! Limits not understood.

•HiO applied to **X-ray** data works well if combined with low-resolution image. Attempts to use the autocorrelation function to define a support are in progress. Some missing data can float freely in the iterations if sufficient constraints compensate the missing information. The resolution is about 10 nm.

• Use of micron-sized SiN windows solves many problems, including how to make and handle an isolated submicron object. (e.g from solution, ink-jet, laser tweezers).